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(54) An epoxy resin composition for encapsulating a semiconductor device.

(57) An epoxy resin composition for encapsulating a semiconductor device comprises (i) an epoxy resin (A) comprising at least one of a bifunctional epoxy resin (a1) having a biphenol skeleton and a bifunctional epoxy resin (a2) having a naphthalene skeleton, (ii) a curing agent (B), and (iii) a filler comprising fused silica (C) consisting of 97 to 50 wt% of crushed fused silica (C1) of a mean particle diameter not more than 10 μm and 3 to 50 wt% of spherical fused silica (C2) of a mean particle diameter not more than 4 μm, wherein the mean particle diameter of the spherical fused silica is smaller than the mean particle diameter of the crushed fused silica, and the amount of the filler is 75 to 90 wt% of the total weight of the composition.

biphenyl ep
naphth ep
nov

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This invention relates to an epoxy resin composition capable of withstanding soldering temperatures.

Epoxy resins have excellent h at resistance, moisture resistance, lectrical charact ristics and adhesion properties, and they can acquire various characteristics on modifying their recipes. Accordingly, therefore, epoxy resins are used in paints, adhesives, and industrial materials such as electrically insulating materials.

As methods of encapsulating electronic circuit parts such as semiconductor devices, there have been proposed a hermetic encapsulating method using metals or ceramics, and a resin encapsulating method using, for example, a phenolic resin, silicone resin or epoxy resin. From the view point of balancing economy, productivity and physical properties, however, the resin encapsulating method using an epoxy resin is mainly adopted.

On the other hand, integration and automated processing have recently been promoted for the step of mounting parts to a circuit board, and a "surface mounting method" in which a semiconductor device is soldered to the surface of a board has been frequently employed in place of the conventional "insertion mounting method" in which lead pins are inserted into holes of a board. Packages are correspondingly in a transient stage of from conventional dual inline package (DIP) to thin-type flat plastic package (FPP) suitable for integrated mounting and surface mounting.

With the transition to the surface mounting method, the soldering process which conventionally has not attracted attention has now come to be a serious problem. According to the conventional pin insertion-mounting method, only a lead part is partially heated during soldering, whereas according to the surface mounting method a package in its entirety is dipped and heated in a heated solvent. Soldering methods for the surface mounting method include the solder-bath dipping method and the solder reflow method in which heating is carried out with, for example, inert-liquid saturated vapor and infrared rays. By any of these methods, a package in its entirety is to be heated at a high temperature of 210-270°C. Accordingly, in a package encapsulated with a conventional encapsulating resin, a problematic cracking of the resin portion occurs at the soldering step, whereby the reliability is lost, and hence, the product obtained is of no practical use.

The occurrence of cracking during the soldering process is regarded as being due to the explosive vaporization and expansion, on heating for soldering, of the moisture absorbed in the time period from procuring to mounting process. As a countermeasure, a method is employed in which a post-cured package is completely dried and enclosed in a hermetically sealed container for shipping.

The improvement of encapsulating resins has been investigated in a wide variety of ways. For example, heat resistance to solder can be improved by a method of adding an epoxy resin having a biphenyl skeleton and a rubber component (JP-A-251419/1988), but this is not sufficient. A method of adding an epoxy resin having a biphenyl skeleton and microparticles, in powder form, of a particle diameter less than 14 um (disclosed in JP-A-87616/1989) does not yield a satisfactory level of heat resistance to solder.

Alternatively, there has been proposed the addition of spherical fused silica microparticles (JP-A-263131/1989), but only the fluidity of the encapsulating resins is improved and the heat resistance to solder is not sufficient.

By employing an epoxy resin composition embodying the present invention, the problem concerning the occurrence of cracking during the soldering process may be minimised or eliminated.

Thus, we find surprisingly that by using an epoxy resin composition embodying the invention a package may be obtained which has excellent heat resistance to solder, excellent reliability after thermal cycles and/or excellent reliability after solder-bath dipping.

The present invention provides an epoxy resin composition for encapsulating a semiconductor device, which composition comprises

(i) an epoxy resin (A) comprising at least one of a bifunctional epoxy resin (a1) having a biphenyl skeleton and a bifunctional epoxy resin (a2) having a naphthalene skeleton,

(ii) a curing agent (B), and

(iii) a filler comprising a fused silica (C) consisting of 97 to 50 wt% of crushed fused silica (C1) of a mean particle diameter not more than 10 μm and 3 to 50 wt% of spherical fused silica (C2) of a mean particle diameter not more than 4 μm, wherein the mean particle diameter of the spherical fused silica smaller than the mean particle diameter of the crushed fused silica, and the amount of the filler is 75 to 90 wt% of the total weight of the composition. The composition may contain additionally a styrene type block copolymer (D), and/or a copolymer (E) of (1) at least one c mpound s lected from ethylene and α-olefin and (2) at least on compound selected from unsaturated carboxylic acid and derivatives thereof.

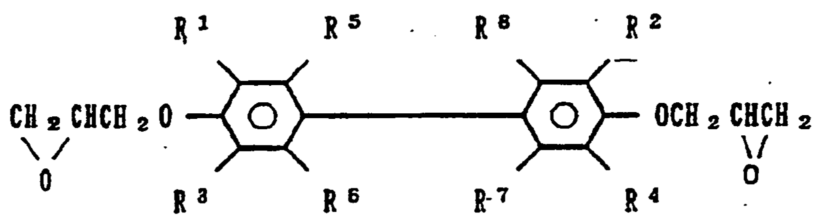
In accordance with the present invention, it is important that an epoxy resin (A) contains as the essential component th reof at least one of a bifunctional epoxy resin (a1) having a biphenyl skeleton and a bifunctional poxy resin (a2) having a naphthalene skeleton, and that a fill r comprising a fused silica (C) is present at 75 to 90 wt% to the total of th comp sition. The fused silica (C) consists of 97 to 50 wt% of crushed fused silica (C1) of a mean particle diameter not more than 10 μ m and 3 to 50 wt% of spherical fused silica (C2) of a mean

particl diameter not more than 4 μ m wherein the mean particl diameter of the spherical fused silica is smaller than the mean particle diameter of the crushed fused silica. Due to the bifonctionality of the epoxy resins (a1) and (a2), the crosslinking density appropriately can be lowered. Biphenyl and naphthyl skeletons with high resistance to heat are present, so that the effect of reducing the water absorption potency of the cured epoxy resin, as well as the effect of making the cured epoxy resin tough at a higher temperature (a solder-treating temperature) can be achieved. The use of the fused silica of a smaller particle diameter, which is preferably uniformly distributed throughout the composition, can prevent the localization of internal stress imposed on the cured epoxy resin. By making the spherical fused silica of a smaller mean particle diameter present among the crushed silica of a small mean particle diameter, the internal stress being imposed on the cured epoxy resin can be reduced more greatly. This has the effect of improving the strength of the cured epoxy resin, in particular the strength at a high temperature (at the solder-treating temperature). In a composition according to th present invention, the effects of the epoxy resin and the silica are uniquely combined in such a way that a synergistic, remarkable improvement in heat resistance to solder, far beyond expectation, may be achieved.

The epoxy resin (A) to be used in accordance with the present invention comprises as the essential component thereof at least one of a bifunctional epoxy resin (a1) having a biphenyl skeleton and a bifunctional epoxy resin (a2) having a naphthalene skeleton.

The effect of preventing the occurrence of cracking during the soldering process cannot be exhibited when the epoxy resins (a1) and (a2) are not present.

The epoxy resin (a1) in a composition of the present invention may be a compound represented by the following formula (I):



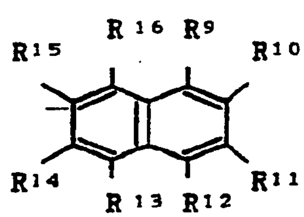
……(I)

wherein R¹ to R⁸ independently represent hydrogen atom, halogen atom or a lower alkyl group having 1 to 4 carbon atoms.

Preferred specific examples of R¹ to R⁸ in the above-mentioned formula (I) are a hydrogen atom, methyl group, ethyl group, propyl group, i-propyl group, n-butyl group, sec-butyl group, tert-butyl group, chlorine atom and bromine atom.

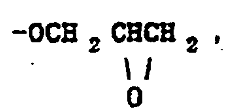
Preferred examples of the epoxy resin (a1) are 4,4'-bis(2,3-epoxypropoxy)biphenyl, 4,4'-bis(2,3-epoxypropoxy)-3,3',5,5'-tetramethylbiphenyl, 4,4'-bis(2,3-epoxypropoxy)-3,3',5,5'-tetramethyl-2-chlorobiph nyl, 4,4'-bis(2,3-epoxypropoxy)-3,3',5,5'-tetramethyl-2-bromobiphenyl, 4,4'-bis(2,3-epoxypropoxy)- 3,3',5,5'-tetraethylbiphenyl, and 4,4'-bis(2,3-epoxypropoxy)-3,3',5,5'-tetrabutylbi-phenyl.

Particularly preferred ewamples are 4, 4'-bis(2,3-epoxypropoxy)biphenyl, and 4,4'-bis(2,3-epoxypropoxy)-3,3',5,5'-tetramethylbiphenyl. In a composition in accordance with the present invention, the epoxy resin (a2) may be a compound represented by the following formula (II):



……(II)

wherein two of R⁹ to R¹⁶ independently represent a group represented by



and those remaining independently represent hydrogen atom, halogen atom or a lower alkyl group having 1 to 4 carbon atoms.

Those among R^9 to R^{16}, excluding the two representing the group

$$-OCH_2CHCH_2\text{ (epoxy: }CH\text{–}CH_2\text{ bridged by }O\text{)},$$

independently represent hydrogen atom, halogen atom or a lower alkyl group having 1 to 4 carbon atoms. Preferred specific example are a hydrogen atom, methyl group, ethyl group, propyl group, i-propyl group. n-butyl group, sec-butyl group, tert-butyl group, chlorine atom and bromine atom.

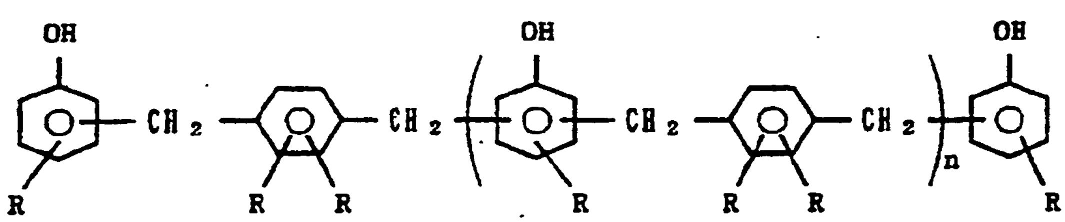
Preferred specific examples of the epoxy resin (a2) are 1,5-di(2,3-epoxypropoxy)naphthalene, 1,5-di(2,3-epoxypropoxy)-7-methylnaphthalene, 1,6-di(2,3-epoxypropoxy)naphthalene, 1,6-di(2,3-epoxypropoxy)-2-methylnaphthalene, 1,6-di(2,3-epoxypropoxy)-8-methylnaphthalene, 1,6-di(2,3-epoxypropoxy)-4,8-dimethylnaphthalene, 2-bromo-1,6-di(2,3-epoxypropoxy)naphthalene, 8-bromo-1,6-di(2,3-epoxypropoxy)naphthalene and 2,7-di(2,3-epoxypropoxy)naphthalene. Particularly preferred examples are 1,5-di(2,3-epoxypropoxy)naphthalene, 1,6-di(2,3-epoxypropoxy)naphthalene and 2,7-di(2,3-epoxypropoxy)naphthalene.

The epoxy resin (A), in a composition of the present invention, may contain at least one epoxy resin other than the epoxy resin (a1) and/or (a2), in combination with the epoxy resin (a1) and/or (a2). Other epoxy resins concurrently usable are cresol-novolac type epoxy resins, phenol-novolac type epoxy resins and various novolac type epoxy resins synthesized from, for example, bisphenol A or resorcine, bisphenol A type epoxy resins, linear aliphatic epoxy resins, alicyclic epoxy resins, heterocyclic epoxy resins and halogenated epoxy resins.

There is no specific limitation as to the ratio of the epoxy resin (a1) and (a2) to be present in the epoxy resin (A), but the effects of the present invention can be achieved only if the epoxy resin (a1) or (a2) is present as the essential component. In order to achieve particularly improved effects, either one or both of the epoxy resins (a1) and (a2) should be present in total in an amount of 50 wt% or more in the epoxy resin (A), preferably 70 wt% or more in the epoxy resin (A).

In a composition in accordance with the present invention, the amount of the epoxy resin (A) present is generally 4 to 20wt%, preferably 6 to 18 wt%, of total weight of the composition.

No specific limitation is imposed on the curing agent (B) in a composition in accordance with the present invention, so long as the agent reacts with the epoxy resin (A) and cures the resin. Specific examples are phenol type curing agents including phenol-novolac resins, cresol-novolac resins, various novolac resins synthesised from, for example, bisphenol A or resorcine, phenol alkylallyic resins represented by the following formula:



wherein n is 0 or an integer; R is a hydrogen atom or a lower alkyl group having 1 to 4 carbon atoms (the Rs are not necessarily identical); trihydroxyphenyl methane; acid anhydrides including maleic anhydride, phthalic anhydride and pyromellitic anhydride; and aromatic amines including metaphenylene diamine, diaminodiphenyl methane and diaminodiphenyl sulfone. For encapsulating a semiconductor device, a phenolic curing agent is preferably used, taking into account the heat resistance, moisture resistance and storage stability th reby achieved; particularly preferable are phenol-novolac resins, phenol alkylallylic resins and trihydroxyphenyl methane. Depending on the use, two or more curing ag nts may be used in combination.

In a composition according to the present invention, the amount of the curing agent (B) mixed into the composition is g nerally 3 to 15 wt%, preferably 4 to 10 wt%,of the total weight of the composition. In order to provide the desired mechanical properties and moisture resistance, amounts of the poxy resin (A) and the curing ag nt (B) compounded are preferably such that the chemical quivalent ratio of the curing agent (B) to th epoxy resin (A) is in th range of 0.7 to 1.3, more preferably in the range of 0.8 to 1.2.

In th pres nt invention, a curing catalyst may be used for promoting the curing reaction between the poxy

resin (A) and the curing agent (B). Any compound capable of promoting the curing reaction can be used in the present invention without specific limitation. For example, there can be included imidazole compounds such as 2-methylimidazole, 2,4-dimethylimidazole; 2-ethyl-4-methylimidazole, 2-phenylimidazol , 2-phenyl-4-methylimidazole, 2-heptadecylimidazole ; tertiary amine compounds such as triethylamine, benzyldimethylamine, α-methylbenzyldimetbylaain , 2- (dimethylaminomethel)phenol, 2,4,6-tris(dimethylaminomethyl)phenol, and 1,8-diazabicyclo(5,4,0)undecene-7 ; organic metal compounds such as zirconium tetramethoxide, zirconium tetrapropoxide, tetrakis(acetylacetonate)zirconium and tri(acetylacetonate)aluminum ; and organic phosphine compounds such as triphenylphosphine, trimethylphosphine, triethylphosphine, tributylphosphine, tri(p-methylphenyl)phosphine, and tri(nonylphenyl)phosphine. From the viewpoint of moistur resistance, an organic phosphine compound is preferable, and triphenylphosphine in particular is preferably used. A combination of two or more of these curing catalysts may be used, depending on the use. Preferably, the curing catalyst is incorporated in an amount of 0.5 to 5 parts by weight per 100 parts by weight of the epoxy resin (A).

In a composition of the present invention, the filler comprises the fused silica (C).

The fused silica (C) in accordance with the present invention consists of 90 to 50 wt% of crushed fused silica of a mean particle diameter not more than 10 μ m and 3 to 50 wt% of spherical fused silica of a mean particle diameter not more than 4 μ m, wherein the mean particle diameter of the spherical fused silica is smaller than the mean particle diameter of the crushed fused silica. Preferably, the fused silica (C) in accordance with the present invention consists of 97 to 60 wt% of crushed fused silica of a mean particle diameter not more than 10 μ m and 3 to 40 wt% of spherical fused silica of a mean particle diameter not more than 4 μ m, wherein the mean particle diameter of the spherical fused silica is smaller than the mean particle diameter of the crushed fused silica. When the ratio of crushed fused silica to spherical fused silica falls outside this range, the composition may not have so excellent a resistance to solder. Crushed fused silica of a mean particle diameter exceeding 10 μ m cannot yield satisfactory heat resistance to solder. There is no specific limitation to the crushed fused silica herein, as long as its mean particle diameter is not more than 10 μ m, but a crushed fused silica of a mean particle diameter of 3 μ m or more and 10 μ m or less is preferably used, from the viewpoint of heat resistance to solder. A crushed fused silica of a mean particle diameter of not less than 3 μ m and less than 7 μ m is specifically preferably used. Provided that the mean particle diameter of crushed fused silica is 10 μ m or less, two or more types of crushed fused silica, with different mean particle diameters, may be us d in combination. The spherical fused silica of a mean particle diameter exceeding 4 μ m cannot yield satisfactory h at resistance to solder. There is no specific limitation to the spherical fused silica, as long as its mean particle diameter is not more than 4 μ m, but a spherical fused silica of a mean particle diameter of 0.1 μ m or more and 4 μ m or less is preferably used, for greater heat resistance to solder. Provided that the mean particle diameter of spherical fused silica is 4 μ m or less two or more types of spherical fused silica, with different mean particle diameters, may be used in combination. The mean particle diameter referred to herein means the particle diameter (median size) at which the cumulative weight reaches 50 wt%. For measuring particle diam ter, a particle diameter distribution measuring method of the laser diffraction type is employed. For laser diffraction type measurement, there is used, for example, a Laser Granulometer Model 715 manufactured by CILAS Co., Ltd. In the fused silica (C), it is also important that the mean particle diameter of spherical fused silica is smaller than the mean particle diameter of the crushed fused silica. When the mean particle diameter of the spherical fused silica is greater than the mean particle diameter of the crushed fused silica, a composition with excellent heat resistance to solder cannot be obtained. The mean particle diameter of the spherical fused silica is smaller than the mean particle diameter of the crushed fused silica. Preferably, the mean particle diameter of spherical fused silica is two-thirds or less of the mean particle diameter of the crushed fused silica, more preferably half or less.

In a composition of the present invention, the amount of the fused silica (C) is preferably at least 80, more preferably at least 90 wt% of the total amount of the filler. When the amount of the fused silica (C) is less than 80 wt% of the total amount of the filler, heat resistance to solder may not be sufficient. The amount of the filler is 75 to 90 wt%, more preferably 77 to 88 wt% of the total amount of the composition. When the amount of the filler is less than 75 wt% or exceeds 90 wt% of the total amount of the composition, heat resistance to solder is not sufficient.

To the epoxy resin composition of the present invention may be added, as additional filler, crystalline silica, calcium carbonate, magnesium carbonate, alumina, magnesia, clay, talc, calcium silicate, titanium oxide, antimony oxide, asbestos or gloss fiber, besides fused silica (C).

In a composition in accordance with th present invention, a polystyren type block copolymer (D) is preferably also included. The polystyrene type block copolymer (D) includes linear, parabolic or branched block copolymers comprising blocks of an aromatic vinyl hydrocarbon polymer having a glass transition temperature of at least 25 °C, preferably at least 50 °C, and blocks of a conjugat d diene polymer having a glass transition

temperature not higher than 0 °C, preferably not higher than -25 °C.

The aromatic vinyl hydrocarbon may be, for example, styrene, a -methylstyrene, o-methylstyrene, p-methylstyrene, 1, 3-demethylstyrene or vinylnaphthalene, and among these, styrene is preferably used.

The conjugated diene may b , for example, butadiene (1, 3-butadiene), isoprene (2-methyl-1,3-butadiene), methylisoprene (2,3-dimethyl-1,3-butadiene) or 1,3-pentadiene, and of these conjugated dienes, butadiene and isoprene are preferably used.

The proportion of the blocks of the aromatic vinyl hydrocarbon, which are blocks of the glass phase, in the block copolymer, is preferably 10 to 50 wt%, and the blocks of the conjugated diene polymer, which are blocks of the rubber phase, is preferably 90 to 50 wt%.

A great number of combinations of the blocks of the glass phase and the blocks of the rubber phas are possible and any of these combinations can be adopted. A diblock copolymer comprising a single block of rubber phase bonded to a single block of glass phase, and a triblock copolymer comprising blocks of the glass phase bonded to both ends of the intermediate block of the rubber phase are preferably used. In this case, the number average molecular weight of the block of the glass phase is preferably 1,000 to 100,000, more preferably 2,000 to 50,000, and the number average molecular weight of the block of the rubber phase is preferably 5,000 to 200,000, more preferably 10,000 to 100,000.

The polystyrene type block copolyner (D) can be prepared by the known living anion polymerization process, but the preparation thereof is not limited to this polymerization process. Thus, alternatively polystyrene type block copolymer (D) can be produced by a cationic polymerization process or a radical polymerization process.

The polystyrene type block copolymer (D) may also be a hydrogenated block copolymer formed by reducing parts of unsaturated bonds of the above-mentioned block copolymer by hydrogenation.

In this case, preferably not more than 25 % of the aromatic double bonds of the blocks of the aromatic vinyl hydrocarbon polymer is hydrogenated, and not less than 80 % of the aliphatic double bonds of the blocks of the conjugated diene polymer is hydrogenated.

Preferred examples of the polystyrene type block copolymer (D) are polystyrene/polybutadiene/polystyrene triblock copolymer(SBS), polystyrene/polyisoprene/polystyrene triblock copolymer(SIS), hydrogenated copolymer of SBS(SEBS), hydrogenated copolymer of SIS, polystyrene/isoprene diblock copolymer and hydrogenated copolymer of the polystyrene/isoprene diblock copolymer (SEP).

The amount of polystyrene type block copolymer (D) incorporated is generally 0.2 to 10 wt%, preferably 0.5 to 5 wt% of total weight of the composition. The effect of improving the heat resistance to solder and reliability on moisture resistance are not sufficient when less than 0. 2 wt% is employed, whereas it is not practical to use an amount exceeding 10 wt% because the molding gets hard due to the lowered fluidity.

When a polystyrene type block copolymer (D) is additionally used in the present invention, heat resistance to solder is thereby further improved, and the reliability after thermal cycling is further improved. The reason is assumed to lie in the synergistic action of the following two effects ;

(1) Polystyrene type block copolymer (D) makes the cured epoxy resin hydrophobic.

(2) Over a wide temperature range, the block of the conjugated diene copolymer in the polystyrene type block copolymer reduces the internal stress generating between semiconductor chips and the cured epoxy resin.

In the present invention, it is preferred to use additionally a copolymer (E) of (1) at least on compound selected from ethylene and α-olefin and (2) at least one compound selected from unsaturated carboxylic acid and derivatives thereof.

The compound selected from ethylene and α-olefin in the copolymer (E) may be, for example, ethylene, propylene, butene-1, pentene-1, 4-methylpentene-1 or octene-1, and of these, ethylene is preferably used. Two or more of ethylene or α-olefin may be concurrently used, depending on the use. The unsaturated carboxylic acid may be, for example, acrylic acid, methacrylic acid, ethyl acrylic acid, crotonic acid, maleic acid, fumaric acid, itaconic acid, citraconic acid or butene dicarboxylic acid. The derivative thereof may be, for example, an alkyl ester, glycidyl ester, acid anhydride or imide thereof. Specific examples are methyl acrylate, ethyl acrylate, propyl acrylate, butyl acrylate, methyl methacrylate, ethyl methacrylate, glycidyl acrylate, glycidyl methacrylate, glycidyl ethyl acrylate, diglycidyl itaconate ester, diglycidyl citraconate ester, diglycidyl butene dicarboxylate ester, monoglycidyl butene dicarboxylate ester, maleic anhydride, itaconic anhydride, citraconic anhydride, maleic imide, N-ph nylmaleic imide, itaconic imide and citraconic imid , and of these, acrylic acid, methacrylic acid, glycidyl acrylate, glycidyl methacrylate,and maleic anhydride are preferably used. These unsaturated carboxylic acids and the derivatives thereof may be used in a combination of two or more.

Having regard to heat resistance to solder and m isture resistance, the copolym rizing amount of a compound selected from unsaturated carboxylic acid and derivatives thereof is pref rably 0. 01 to 50 wt%.

Preferably, the melt index of the copolymer(E), measured according to ASTM-D1238, is 0.1 to 5,000, more preferably 1 to 3,000, having regard to moldability and heat resistance to solder.

Having regard to resistance to solder and moisture resistance, the added amount of the copolymer of (E) is g nerally 0.1 to 10 wt%, preferably 0.5 to 5 wt%, more pr ferably 1 to 4 wt% of the total w ight of th composition.

The copolymer (E) may be preliminarily mad into a powder, by means of grinding, crosslinking, and other means, in accordance with th present inventi n.

The copolymer (E) can be compounded by appropriate procedures. For example, the copolymer may be preliminarily melt mixed with the epoxy resin (A) or the curing agent (B) followed by addition of other components or the copolymer may be compounded simultaneously with the epoxy resin (A), the curing agent (B) and other components.

When the copolymer of (E) is used in the present invention, heat resistance to solder is thereby further improved and the reliability after dipping in a solder bath is much more improved. The reason is assumed to be due to the synergistic action of the following two effects ;

(1) The copolymer makes the cured epoxy resin hydrophobic.

(2) Part of the unsaturated carboxylic acid or a derivative thereof in the copolymer reacts with the epoxy resin or the curing agent to render the cured epoxy resin tough.

Having regard to reliability, preferably the fused silica (C) and any other filler component is prelliminary surface treated with a coupling agent including silane coupling agent and titanate coupling agent. Preferably, silane coupling agents such as epoxysilane, aminosilane, and mercaptosilane, are preferably used.

A flame retardant such as a halogenated epoxy resin or phosphorus compounds, a flame retardant assistant such as antimony trioxide, a colorant such as carbon black or iron oxide, an elastomer such as silicone rubber, modified nitrile rubber or modified polybutadiene rubber, a thermoplastic resin such as p lyethylene, a release agent such as long-chain fatty acid, metal salt of long-chain fatty acid, ester of long-chain fatty acid, amide of long-chain fatty acid, paraffin wax or modiofied silicone oil, and a crosslinking agent such as an organic peroxide can be added to the epoxy resin composition of the present invention.

The epoxy resin composition of the present invention is preferably melt-kneaded. For example, the poxy resin composition can be prepared by carrying out the melt-kneading according to a known kneading method using a Banbury mixer, a kneader, a roll, a single-screw or twin-screw extruder or a cokneader.
Embodiments of the present invention will now be described in more detail with reference to the following examples.

Examples 1 to 20

Using fused silica of each of the compositions shown in Table 1, blending of the reagents was carried out at their mixing ratios shown in Table 2, by using a mixer. The blend was melt-kneaded using a twin-screw extruder having a barrel-preset temperature maintained at 90°C, and then cooled and pulverized to prepare an epoxy resin composition.

Using the composition, a test device was molded according to the low-pressure transfer molding m thod to evaluate the heat resistance to solder under the conditions described below.

Evaluation of heat resistance to solder :

Thirty-two each of 80-pin QFP (package size, 17 x 17x 1.7 mm ; silicone chip size, 9 x 9 x 0.5 mm) were molded and cured at 180°C for 5 hours, followed by humidification at 85°C/85 % RH for 50 hours. Then, ach of sixteen 80-pin QFP was dipped into a solder bath heated at 260°C for 10 seconds, while each of another sixteen 80-pin QFP , was placed into a VPS (vapor phase solder reflow) furnace heated at 215 °C for 90 seconds. Those QFP with occurrence of cracking were judged defective.

The results are shown in Table 3.

As is shown in Table 3, the epoxy resin compositions of the present invention (Examples 1 to 20) have excellent heat resistance to solder.

Table 1 Compositions of fused silica

	Crushed fused silica		Spherical fused silica		Crushed fused silica/Spherical fused silica
	Ratio by weight*1 (I/II/III/IV/V)	Mean particle diameter (μm)	Ratio by weight *2 (VI/VII/VIII)	Mean particle diameter (μm)	Ratio by weight
Example 1	0/100/0/0/0	5.3	100/0/0	0.2	95/5
Example 2	0/100/0/0/0	5.3	0/100/0	2.1	95/5
Example 3	0/100/0/0/0	5.3	0/100/0	2.1	95/5
Example 4	100/0/0/0/0	3.4	100/0/0	0.2	90/10
Example 5	0/0/100/0/0	6.5	0/100/0	2.1	90/10
Example 6	0/0/100/0/0	6.5	0/100/0	2.1	90/10
Example 7	100/0/0/0/0	3.4	100/0/0	0.2	80/20
Example 8	0/0/100/0/0	6.5	0/100/0	2.1	80/20
Example 9	0/0/100/0/0	6.5	0/100/0	2.1	80/20
Example 10	0/0/100/0/0	6.5	0/100/0	2.1	90/10
Example 11	0/0/0/100/0	8.9	100/0/0	0.2	90/10
Example 12	0/0/70/0/30	9.2	0/100/0	2.1	90/10
Example 13	0/100/0/0/0	5.3	100/0/0	0.2	80/20
Example 14	0/0/0/100/0	8.9	0/70/30	3.6	80/20
Example 15	0/100/0/0/0	5.3	100/0/0	0.2	80/20
Example 16	50/0/0/50/0	6.3	50/50/0	0.9	80/20
Example 17	0/0/100/0/0	6.5	0/100/0	2.1	70/30
Example 18	0/0/100/0/0	6.5	0/100/0	2.1	70/30
Example 19	50/0/05/0/0	6.3	50/50/0	0.9	80/20
Example 20	0/0/0/100/0	8.9	0/100/0	2.1	70/30

*1 Mean particle diam t r of crushed fused silica (μm) [I:3.4, II:5.3, III:6.5, IV:8.9, V:14.0]
*2 Mean particle diamet r of spherical fused silica (μm) [VI:0.2, VII:2.1, VIII:6.5]

Table 2 Epoxy Resin Compositions (wt%)

	Epoxy resin			Curing agent		Curing catalyst
	4,4'-Bis(2,3-epoxypropoxy)-3,3',5,5'-tetramethylbiphenyl	1,6-Di(2,3-epoxypropoxy)-naphthalene	Ortho-cresol novolac type epoxy resin of an epoxy equivalent of 200	Phenol novolac resin of a hydroxyl group equivalent of 107	Phenol alkylallylic resin of a hydroxyl group equivalent of 173	Triphenylphosphine
Example 1	9.4	0.0	0.0	6.3	0.0	0.2
Example 2	9.4	0.0	0.0	6.3	0.0	0.2
Example 3	0.0	8.5	0.0	7.2	0.0	0.2
Example 4	8.6	0.0	0.0	5.9	0.0	0.2
Example 5	8.1	0.0	0.0	5.6	0.0	0.2
Example 6	0.0	7.2	0.0	6.5	0.0	0.2
Example 7	8.1	0.0	0.0	5.6	0.0	0.2
Example 8	6.7	0.0	1.7	5.3	0.0	0.2
Example 9	0.0	5.0	3.3	5.4	0.0	0.2
Example 10	0.0	6.0	0.0	0.0	7.7	0.2
Example 11	7.6	0.0	0.0	5.1	0.0	0.2
Example 12	7.6	0.0	0.0	5.1	0.0	0.2
Example 13	7.6	0.0	0.0	5.1	0.0	0.2
Example 14	7.6	0.0	0.0	5.1	0.0	0.2
Example 15	0.0	6.7	0.0	6.0	0.0	0.2
Example 16	7.1	0.0	0.0	4.6	0.0	0.2
Example 17	7.1	0.0	0.0	4.6	0.0	0.2
Example 18	0.0	6.1	0.0	5.6	0.0	0.2
Example 19	3.3	3.3	0.0	5.1	0.0	0.2
Example 20	6.0	0.0	0.0	3.8	0.0	0.1

Table 2 (continued) Epoxy Resin Compositions (wt%)

	Fused silica in Table 1	Silane coupling agent	Flame Retardant	Flame retardant assistant	Colorant	Release agent
		γ-Glycidoxy-propyltri-methoxysilane	Brominated phenol novolac type epoxy resin with an epoxy equivalent of 270 and a total bromine content of 36wt%	Antimony trioxide	Carbon black	Carnauba wax
Exampl 1	79	0.7	2.3	1.5	0.3	0.3
Example 2	79	0.7	2.3	1.5	0.3	0.3
Example 3	79	0.7	2.3	1.5	0.3	0.3
Example 4	80	0.7	2.3	1.5	0.3	0.3
Exampl 5	81	0.7	2.3	1.5	0.3	0.3
Example 6	81	0.7	2.3	1.5	0.3	0.3
Example 7	81	0.7	2.3	1.5	0.3	0.3
Example 8	81	0.7	2.3	1.5	0.3	0.3
Example 9	81	0.7	2.3	1.5	0.3	0.3
Exampl 10	81	0.7	2.3	1.5	0.3	0.3
Example 11	82	0.7	2.3	1.5	0.3	0.3
Example 12	82	0.7	2.3	1.5	0.3	0.3
Example 13	82	0.7	2.3	1.5	0.3	0.3
Example 14	82	0.7	2.3	1.5	0.3	0.3
Example 15	82	0.7	2.3	1.5	0.3	0.3
Example 16	83	0.7	2.3	1.5	0.3	0.3
Example 17	83	0.7	2.3	1.5	0.3	0.3
Example 18	83	0.7	2.3	1.5	0.3	0.3
Example 19	83	0.7	2.3	1.5	0.3	0.3
Example 20	85	0.7	2.3	1.5	0.3	0.3

Table 3 Results of evaluation

	Heat resistance to solder	
	Dipping in solder at 260℃ (Fraction defective)	Solder reflow at 215℃ (Fraction defective)
Example 1	2/16	2/16
Example 2	2/16	2/16
Example 3	3/16	0/16
Example 4	0/16	2/16
Example 5	0/16	0/16
Example 6	0/16	0/16
Example 7	0/16	2/16
Example 8	4/16	2/16
Example 9	6/16	2/16
Example 10	0/16	0/16
Example 11	0/16	0/16
Example 12	2/16	1/16
Example 13	0/16	0/16
Example 14	3/16	0/16
Example 15	0/16	0/16
Example 16	1/16	0/16
Example 17	0/16	0/16
Example 18	2/16	0/16
Example 19	0/16	0/16
Example 20	4/16	0/16

Comparative Examples 1 to 10

Using fused silica of each of the compositions shown in Table 4, blending of the reagents was carried out at their mixing ratios shown in Table 5, by using a mixer. Epoxy resin compositions were produced as in Examples 1 to 20, and the compositions were subjected to the evaluation of heat resistance to solder.

The results are shown in Table 6 and Table 7.

As is shown in Table 6, all of the compositions with the incorporated amounts of fused silica being outside the range of the present invention (Comparative Examples 1 and 10), the compositions not containing the epoxy resin of the present invention (Comparative Examples 2 and 7), the compositions with the incorporated amounts of spherical fused silica being outside the range of the present invention (Comparative Examples 3, 4 and 9), the composition with a mean particle diameter of spherical fused silica being greater than the mean particle diameter of crushed fused silica (Comparative Example 5), and the compositions with the mean particle diameter of crushed fused silica or spherical fused silica being outside the range of the present invention (Comparative Examples 6 and 8), have much poorer heat resistance to solder in contrast to the epoxy resin compositions of the present invention.

As is shown in Table 7, more excellent heat resistance to solder can be obtained even at more strict conditions for evaluating heat resistance to solder, when, in accordance with the invention, the mean particle diameter of crushed fused silica is les than 7 μm (Examples 5, 7, 10, 13 and 15) than when the mean particle diameter of crushed fused silica is 7 to 10 μm (Examples 11, 12 and 14).

Table 4 Compositions of fused silica

	Crushed fused silica		Spherical fused silica		Crushed fused silica/Spherical fused silica
	Ratio by weight*1 (I/II/III/IV/V)	Mean particle diameter (μm)	Ratio by weight*1 (VI/VII/VIII)	Mean particle diameter (μm)	Ratio by weight
Comparative Example 1	0/100/0/0/0	5.3	0/100/0	2.1	95/5
Comparative Example 2	0/0/0/100/0	8.9	100/0/0	0.2	90/10
Comparative Example 3	0/100/0/0/0	5.3	0/0/0	—	100/0
Comparative Example 4	0/100/0/0/0	5.3	0/0/0	—	100/0
Comparative Example 5	100/0/0/0/0	3.4	0/70/30	3.6	90/10
Comparative Example 6	0/0/0/0/100	14.0	0/100/0	2.1	90/10
Comparative Example 7	0/0/100/0/0	6.5	0/100/0	2.1	80/20
Comparative Example 8	0/0/0/100/0	8.9	0/0/100	6.5	90/10
Comparative Example 9	0/0/100/0/0	6.5	0/100/0	2.1	40/60
Comparative Example 10	0/0/100/0/0	6.5	0/100/0	2.1	60/40

*1 Mean particle diameter of crush d fused silica (μm) [I:3.4, II:5.3, III:6.5, IV:8.9, V:14.0]

*2 Mean particle diameter of spherical fused silica (μm) [VI:0.2, VII:2.1, VIII:6.5]

Table 5 Epoxy Resin Compositions (wt%)

	Epoxy resin			Curing agent	Curing catalyst
	4,4'-Bis(2,3-epoxypropoxy)-3,3',5,5'-tetramethyl biphenyl	1,6-Di(2,3-epoxypropoxy)-naphthalene	Ortho-cresol novolac type epoxy resin of an epoxy equivalent of 200	Phenol novolac resin of a hydroxyl group equivalent of 107	Triphenyl-phosphine
Comparative Example 1	12.9	0.0	0.0	8.7	0.3
Comparative Example 2	0.0	0.0	11.0	6.7	0.2
Comparative Example 3	9.4	0.0	0.0	6.3	0.2
Comparative Example 4	0.0	8.5	0.0	7.2	0.2
Comparative Example 5	8.8	0.0	0.0	5.9	0.2
Comparative Example 6	8.1	0.0	0.0	5.6	0.2
Comparative Example 7	0.0	0.0	8.4	5.3	0.2
Comparative Example 8	7.6	0.0	0.0	5.1	0.2
Comparative Example 9	6.4	0.0	0.0	4.4	0.1
Comparative Example 10	1.8	0.0	0.0	2.0	0.1

Table 5 (continued) Epoxy Resin Compositions (wt%)

	Fused silica in Table 4	Silane coupling agent	Flame Retardant	Flame retardant assistant	Colorant	Releas agent
		γ-Glycidoxy-propyltri-methoxysilane	Brominated phenol novolac type epoxy resin with an epoxy equivalent of 270 and a total bromine content of 36wt%	Antimony trioxide	Carbon black	Carnauba wax
Comparative Example 1	73	0.7	2.3	1.5	0.3	0.3
C mparative Example 2	77	0.7	2.3	1.5	0.3	0.3
Comparative Example 3	79	0.7	2.3	1.5	0.3	0.3
Comparative Example 4	79	0.7	2.3	1.5	0.3	0.3
Comparative Example 5	80	0.7	2.3	1.5	0.3	0.3
Comparative Example 6	81	0.7	2.3	1.5	0.3	0.3
Comparative Example 7	81	0.7	2.3	1.5	0.3	0.3
Comparative Example 8	82	0.7	2.3	1.5	0.3	0.3
Comparative Example 9	84	0.7	2.3	1.5	0.3	0.3
Comparative Example 10	91	0.7	2.3	1.5	0.3	0.3

Table 6 Results of evaluation

	Heat resistance to solder	
	Dipping in solder at 260℃ (Fraction defective)	Solder reflow at 215℃ (Fraction defective)
Comparative Example 1	16/16	16/16
Comparative Example 2	16/16	16/16
Comparative Example 3	11/16	10/16
Comparative Example 4	16/16	12/16
Comparative Example 5	9/16	15/16
Comparative Example 6	16/16	9/16
Comparative Example 7	Melt-kneading was impossible ; evaluation impossible	
Comparative Example 8	14/16	11/16
Comparative Example 9	16/16	12/16
Comparative Example 10	Melt-kneading was impossible ; evaluation impossible	

Table 7 Results of evaluation

	Heat resistance to solder Dipping in solder at 260℃	
	After 50-hour humidification (Fraction defective)	After 75-hour humidification (Fraction defective)
Example 5	0/16	0/16
Example 7	0/16	0/16
Example 10	0/16	0/16
Example 11	0/16	4/16
Example 12	2/16	6/16
Example 13	0/16	0/16
Example 14	3/16	6/16
Example 15	0/16	0/16
Comparative Example 2	16/16	16/16
Comparative Example 6	16/16	16/16
Comparative Example 8	14/16	16/16

Examples 21 to 38, Comparative Examples 11 to 16

Using the styrene type block copolymers each shown in Table 8 and the fused silica of each of th compositions shown in Table 9, blending of the reagents was carried out at their mixing ratios shown in Table 10, by using a mixer. Epoxy resin compositions were produced as in Examples 1 to 20.

Using the compositions, test devices were molded according to the low-pressure transfer molding method to evaluate the heat resistance to solder and reliability on moisture resistance after thermal cycling.

Evaluation of heat resistance to solder :

Sixteen 80-pin QFP were molded and post cured at 180°C for 5 hours, followed by humidification at 85°C/85 % RH for 48 hours, which were then dipped into a solder bath heated at 260°C for 10 seconds. Those QFP with occurrence of cracking were judged defective.

Evaluation of reliability on moisture resistance after thermal cycling :

Twenty 16-pin DIP (package size, 19 x 6 x 3 mm) mounting a test element with aluminum wiring were molded and cured at 180°C for 5 hours, followed by 100-time repetition of the thermal cycle from -55°C to 150°C which were then subjected to PCT under the conditions of 143°C /100 % RH. Then, the lifetime of the properties was determined in Weibull distribution.

The results are shown in Table 11.

As shown in Table 11, the epoxy resin compositions with the styrene type block copolymers added, in accordance with the present , invention (Examples 21 to 34), have improved heat resistance to solder together with considerably improved reliability on moisture resistance after thermal cycling, compared with those compositions without styrene type block copolymers added (Examples 35 to 38).

All of the composition with the mean particle diameter of spherical fused silica greater than the size of crushed fused silica (Conparative Example 11), the composition not containing the spherical fused silica (Comparative Example 12), the compositions not containing the epoxy resin composition of the present invention (Comparative Examples 13 and 14), and the compositions of the mean particle diameter of crushed or spherical fused silica being outside the range of the present invention (Comparative Examples 15 and 16), even though the above compositions all contain styrene type block copolymers, have much poorer heat resistance to solder and reliability on moisture resistance after thermal cycling, in contrast to the epoxy resin compositions of the present invention.

Table 8

Polystyrene type block copolymer	Copolymerized composition	Polymerized ratio of copolymers		Solution viscosity (cps) 25 ℃
		Styrene	Butadiene or isoprene	
I	Polystyrene/polybutadiene/polystyrene triblock copolymer	40	60	2,500 *1
II	Hydrogenated polystyrene/polybutadiene/polystyrene triblock copolymer	29	71	550 *2
III	Hydrogenated polystyrene/polybutadiene/polystyrene triblock copolymer	13	87	1,100 *2
IV	Polystyrene/polyisoprene/polystyrene triblock copolymer	21	79	1,300 *1
V	Hydrogenated polystyrene/polyisoprene diblock copolymer	37	63	1,260 *3

*1 25 wt% toluene solution

*2 20 wt% toluene solution

*3 15 wt% toluene soluti n

Table 9 Compositions of fused silica

	Crushed fused silica		Spherical fused silica		Crushed fused silica/Spherical fused silica
	Ratio by weight*1 (I/II/III/IV/V)	Mean particle diameter (μm)	Ratio by weight*2 (VI/VII/VIII)	Mean particle diameter (μm)	Ratio by weight
Example 21	0/0/100/0/0	6.5	100/0/0	0.2	95/5
Example 22	30/0/70/0/0	6.0	50/50/0	0.9	95/5
Example 23	100/0/0/0/0	3.4	100/0/0	0.2	90/10
Example 24	0/0/100/0/0	6.5	0/100/0	2.1	80/20
Example 25	0/0/100/0/0	6.5	0/100/0	2.1	80/20
Example 26	0/0/0/100/0	8.9	0/100/0	2.1	70/30
Example 27	0/0/0/100/0	8.9	100/0/0	0.2	90/10
Example 28	0/0/0/100/0	8.9	100/0/0	0.2	90/10
Example 29	0/0/0/100/0	8.9	0/70/30	3.6	90/10
Example 30	0/0/100/0/0	6.5	0/100/0	2.1	70/30
Example 31	0/0/0/100/0	8.9	0/100/0	2.1	80/20
Example 32	0/0/0/100/0	8.9	0/100/0	2.1	80/20
Example 33	0/0/100/0/0	6.5	0/100/0	2.1	80/20
Example 34	0/0/100/0/0	6.5	0/100/0	2.1	80/20
Comparative Example 11	100/0/0/0/0	3.4	0/70/30	3.6	90/10
Comparative Example 12	0/0/0/100/0	8.9	0/0/0	-	100/0
Comparative Example 13	0/0/0/100/0	8.9	100/0/0	0.2	90/10
Comparative Example 14	0/0/0/100/0	8.9	0/100/0	2.1	70/30
Comparative Example 15	0/0/0/0/100	14.0	0/100/0	2.1	80/20
Comparative Example 16	0/0/0/100/0	8.9	0/0/100	6.5	80/20
Example 35	0/0/100/0/0	6.5	0/100/0	2.1	80/20
Example 36	0/0/0/100/0	8.9	100/0/0	0.2	90/10
Example 37	0/0/100/0/0	6.5	0/100/0	2.1	70/30
Example 38	0/0/0/100/0	8.9	0/100/0	2.1	80/20

*1 Mean particle diameter of crushed fused silica (μm) [I:3.4, II:5.3, III:6.5, IV:8.9, V:14.0]
*2 Mean particle diameter of spherical fused silica (μm) [VI:0.2, VII:2.1, VIII:6.5]

Table 10 Epoxy Resin Compositions (wt%)

	Epoxy resin			Curing agent	Curing catalyst	Fused silica in Table 9
	4,4'-Bis(2,3-epoxypropoxy)-3,3',5,5'-tetramethylbiphenyl	1,6-Di(2,3-epoxypropoxy)-naphthalene	Ortho-cresol novolac type epoxy resin of an epoxy equivalent of 200	Phenol novolac resin of a hydroxyl group equivalent of 107	Triphenylphosphine	
Example 21	9.8	0.0	0.0	5.9	0.2	78
Example 22	10.4	0.0	0.0	6.3	0.2	78
Example 23	10.4	0.0	0.0	6.3	0.2	78
Example 24	5.9	0.0	3.9	5.9	0.2	78
Example 25	0.0	5.6	3.7	6.4	0.2	78
Example 26	7.9	0.0	0.0	4.8	0.2	79
Example 27	9.1	0.0	0.0	5.6	0.2	79
Example 28	4.4	4.4	0.0	5.9	0.2	79
Example 29	0.0	7.7	0.0	6.0	0.2	80
Example 30	7.9	0.0	0.0	4.8	0.2	80
Example 31	7.9	0.0	0.0	4.8	0.2	81
Example 32	0.0	7.2	0.0	5.5	0.2	81
Example 33	0.0	6.6	0.0	5.1	0.2	82
Example 34	7.2	0.0	0.0	4.5	0.2	83
Comparative Example 11	10.4	0.0	0.0	6.3	0.2	78
Comparative Example 12	9.1	0.0	0.0	5.6	0.2	79
Comparative Example 13	0.0	0.0	9.3	5.4	0.2	79
Comparative Example 14	0.0	0.0	8.1	4.6	0.2	79
Comparative Example 15	0.0	7.2	0.0	5.5	0.2	81
Comparative Example 16	7.9	0.0	0.0	4.8	0.2	81
Example 35	0.0	6.3	4.2	7.2	0.2	78
Example 36	10.4	0.0	0.0	6.3	0.2	79
Example 37	9.8	0.0	0.0	5.9	0.2	80
Example 38	0.0	8.3	0.0	6.4	0.2	81

Table 10 (continued) Epoxy Resin Compositions (wt%)

	Silane coupling agent	Flame Retardant	Flame retardant assistant	Colorant	Release agent	Styrene type block copolymer of Table 8
	γ-Glycidoxy-propyltri-methoxysilane	Tetrabromo-bisphenol A type epoxy resin with an epoxy equivalent of 400 and a total bromine content of 49wt%	Antimony trioxide	Carbon black	Carnauba wax	Type/Quantity
Example 21	0.5	1.5	1.5	0.3	0.3	I : 2.0
Example 22	0.5	1.5	1.5	0.3	0.3	V : 1.0
Example 23	0.5	1.5	1.5	0.3	0.3	IV : 1.0
Example 24	0.5	1.5	1.5	0.3	0.3	V : 2.0
Example 25	0.5	1.5	1.5	0.3	0.3	V : 2.0
Example 26	0.5	1.5	1.5	0.3	0.3	II : 4.0
Example 27	0.5	1.5	1.5	0.3	0.3	III: 2.0
Example 28	0.5	1.5	1.5	0.3	0.3	III: 2.0
Example 29	0.5	1.5	1.5	0.3	0.3	III: 2.0
Example 30	0.5	1.5	1.5	0.3	0.3	III: 3.0
Example 31	0.5	1.5	1.5	0.3	0.3	V : 2.0
Example 32	0.5	1.5	1.5	0.3	0.3	V : 2.0
Example 33	0.5	1.5	1.5	0.3	0.3	II : 2.0
Example 34	0.5	1.5	1.5	0.3	0.3	II : 1.0
Comparative Example 11	0.5	1.5	1.5	0.3	0.3	IV : 1.0
Comparative Example 12	0.5	1.5	1.5	0.3	0.3	III: 2.0
Comparative Example 13	0.5	1.5	1.5	0.3	0.3	III: 2.0
Comparative Example 14	0.5	1.5	1.5	0.3	0.3	II : 4.0
Comparative Example 15	0.5	1.5	1.5	0.3	0.3	V : 2.0
Comparative Example 16	0.5	1.5	1.5	0.3	0.3	V 2.0
Example 35	0.5	1.5	1.5	0.3	0.3	0.0
Example 36	0.5	1.5	1.5	0.3	0.3	0.0
Example 37	0.5	1.5	1.5	0.3	0.3	0.0
Example 38	0.5	1.5	1.5	0.3	0.3	0.0

Table 11 Results of evaluation

	Heat resistance of solder Dipping in solder at 260℃ (Fraction defective)	Reliability on moisture resistance PCT after thermal cycling (hr)
Example 21	0/16	300
Example 22	0/16	310
Example 23	0/16	300
Example 24	1/16	290
Example 25	1/16	290
Example 26	1/16	320
Example 27	0/16	330
Example 28	0/16	300
Example 29	1/16	280
Example 30	0/16	310
Example 31	0/16	330
Example 32	0/16	310
Example 33	0/16	300
Example 34	0/16	280
Comparative Example 11	13/16	150
Comparative Example 12	14/16	140
Comparative Example 13	16/16	140
Comparative Example 14	Melt-kneading was impossible ; evaluation impossible	
Comparative Example 15	16/16	90
Comparative Example 16	16/16	110
Example 35	5/16	220
Example 36	2/16	210
Example 37	2/16	210
Exampl 38	1/16	200

Examples 39 to 55, comparative Examples 17 to 22

Using the copolym rs (E) shown in Table 12, and the fused silica with the compositions shown in Table 13, blending of the reag nts was carried out at their mixing ratios shown in Table 14, by using a mixer, to produce epoxy resin compositions as in Examples 1 to 20.

Using the compositions, test devices were molded according to the low-pressure transfer molding method, which were then subjected to the evaluation of heat resistance to solder and reliability on moisture resistance after dipping in solder.

Evaluation of heat resistance to solder :

Sixteen 80-pin QFP were molded and cured at 180°C for 5 hours, followed by humidification at 85°C/85 % RH for 48 hours, which were then dipped into a solder bath heated at 260°C for, 10 seconds. Those QFP with occurrence of cracking were judged defective.

Evaluation of reliability on moisture resistance after dipping in solder :

A test element with aluminum wiring was mounted on an 80-pin QFP and molded. The resulting test device was cured at 180°C for 5 hours, followed by humidification at 85°C/85 % RH for 48 hours, which was then dipped in a solder bath heated at 260°C for 10 seconds. The test device after dipping in solder was subjected to PCT under the conditions of 143°C/100 % RH, whether or not cracking occurred in the test device. Then, th lifetime of the properties was determined in Weibull distribution.

The results are shown in Table 15 and Table 16.

As shown in Table 15, the epoxy resin compositions with the copolymers (E) being added, in accordance with the present invention (Examples 39 to 51), have improved heat resistance to solder together with considerably-improved reliability on moisture resistance after solder dipping, compared with those compositions without copolymers (E) added (Examples 52 to 55).

All of the compositions not containing the epoxy resin composition of the present invention (Comparative Examples 17 and 21), the composition with the mean particle diameter of spherical fused silica gr ater than the size of crushed fused silica (Comparative Example 18), the compositions of the mean particle diameter of crushed or spherical fused silica being outside the range of the present invention (Comparative Examples 19 and 20), and the composition in which the ratio of spherical fused silica used is outside the range of the present invention (Comparative Example 22), even though the above compositions all contain the copolymers (E), have much poorer heat resistance to solder and reliability on moisture resistance after solder dipping.

Even when using 28-pin SOP instead of 80-pin QFP as test device and changing the testing condition as follows, the epoxy resin compositions of the present invention have been found to have excellent heat resistance to solder and reliability on moisture resistance after solder dipping, as is shown in Table 16.

Evaluation of heat resistance to solder :

A test element with aluminum wiring war mounted on a 28-pin SOP and molded. The resulting test device was cured at 180°C for 5 hours, followed by humidification at 85°C/85 % RH for 72 hours, and was then dipped in a solder bath heated at 260°C for 10 seconds. Those SOP with the occurrence of cracking were judged defective.

Evaluation of reliability on moisture resistance after dupping in solder :

SOP after the evaluation of heat resistance of solder was subjected to PCT under the conditions of 121°C/100 % RH whether or not cracking occurred therein. Then, the time at which the cumulative failure rate reached 50% was determined.

Table 12

Symbols	Copolymer (E)		
	Copolymerized composition	Ratio by weight	Melt index (g/10min)
I	Ethylene/ethyl acrylate	75/25	50
II	Ethylene/acrylic acid	95/5	20
III	Ethylene/ethyl acrylate/maleic anhydride	68/30/2	40
IV	Ethylene/methyl methacrylate	85/15	300
V	Ethylene/glycidyl methacrylate	90/10	10

Table 13 Compositions of fused silica

	Crushed fused silica		Spherical fused silica		Crushed fused silica/Spherical fused silica
	Ratio by weight*1 (I/II/III/IV/V)	Mean particle diameter (μm)	Ratio by weight*2 (VI/VII/VIII)	Mean particle diameter (μm)	Ratio by weight
Example 39	0/100/0/0/0	5.3	100/0/0	0.2	95/5
Example 40	0/100/0/0/0	5.3	0/100/0	2.1	95/5
Example 41	100/0/0/0/0	3.4	100/0/0	0.2	90/10
Example 42	0/0/100/0/0	6.5	0/100/0	2.1	90/10
Example 43	0/0/0/100/0	8.9	0/70/30	3.6	80/20
Example 44	0/0/100/0/0	6.5	0/100/0	2.1	90/10
Example 45	0/0/100/0/0	6.5	0/100/0	2.1	80/20
Example 46	0/0/100/0/0	6.5	0/100/0	2.1	80/20
Example 47	0/0/100/0/0	6.5	0/100/0	2.1	70/30
Example 48	0/0/0/100/0	8.9	0/100/0	2.1	70/30
Example 49	50/0/0/50/0	6.3	50/50/0	0.9	80/20
Example 50	0/0/100/0/0	6.5	0/100/0	2.1	70/30
Example 51	0/0/0/100/0	8.9	0/70/30	3.6	70/30
Comparative Example 17	0/0/0/100/0	8.9	0/100/0	2.1	70/30
Comparative Example 18	100/0/0/0/0	3.4	0/70/30	3.6	80/20
Comparative Example 19	0/0/0/100/0	8.9	0/0/100	6.5	80/20
Comparative Example 20	0/0/0/0/100	14.0	0/100/0	2.1	70/30
Comparative Example 21	0/0/0/100/0	8.9	0/100/0	2.1	70/30
Comparative Example 22	0/0/100/0/0	6.5	0/100/0	2.1	40/60
Example 52	0/0/100/0/0	6.5	0/100/0	2.1	90/10
Example 53	0/0/100/0/0	6.5	0/100/0	2.1	80/20
Example 54	0/0/100/0/0	6.5	0/100/0	2.1	70/30
Example 55	0/0/0/100/0	8.9	0/100/0	2.1	70/30

*1 Mean particle diameter of crushed fused silica (μm) [I:3.4, II:5.3, III:6.5, IV:8.9, V:14.0]
*2 Mean particle diameter of spherical fused silica (μm) [VI:0.2, VII:2.1, VIII:6.5]

Table 14 Epoxy Resin Compositions (wt%)

	Epoxy resin			Curing agent	Curing catalyst	Fused silica in Table 13
	4,4'-Bis(2,3-epoxypropoxy)-3,3',5,5'-tetramethyl-biphenyl	1,6-Di(2,3-epoxypropoxy)-naphthalene	Ortho-cresol novolac type epoxy resin of an epoxy equivalent of 200	Phenol novolac resin of a hydroxyl group equivalent of 107	Triphenyl-phosphine	
Example 39	9.2	0.0	0.0	6.0	0.2	78
Example 40	9.2	0.0	0.0	6.0	0.2	79
Example 41	8.5	0.0	0.0	5.7	0.2	79
Example 42	7.9	0.0	0.0	5.3	0.2	80
Example 43	7.2	0.0	0.0	5.0	0.2	80
Example 44	0.0	7.0	0.0	6.2	0.2	80
Example 45	6.4	0.0	1.6	5.2	0.2	81
Example 46	0.0	6.1	1.5	5.6	0.2	81
Example 47	7.2	0.0	0.0	5.0	0.2	81
Example 48	6.0	0.0	0.0	4.2	0.2	81
Example 49	3.1	3.1	0.0	5.0	0.2	82
Example 50	0.0	5.5	0.0	4.8	0.2	83
Example 51	4.7	0.0	0.0	3.5	0.2	86
Comparative Example 17	0.0	0.0	8.7	5.5	0.2	77
Comparative Example 18	7.2	0.0	0.0	5.0	0.2	80
Comparative Example 19	7.2	0.0	0.0	5.0	0.2	80
Comparative Example 20	7.2	0.0	0.0	5.0	0.2	81
Comparative Example 21	0.0	0.0	6.2	4.0	0.2	81
Comparative Example 22	0.0	5.5	0.0	4.8	0.2	83
Example 52	0.0	8.3	0.0	6.9	0.2	80
Example 53	0.0	6.5	1.6	6.1	0.2	81
Example 54	8.5	0.0	0.0	5.7	0.2	81
Example 55	8.5	0.0	0.0	5.7	0.2	81

Table 14 (continued) Epoxy Resin Compositions (wt%)

	Silane coupling agent	Flame Retardant	Flame retardant assistant	Colorant	Release agent	Copolymer of Table 12
	γ-Glycidoxy propyltri-methoxysilane	Brominated phenol novolac type epoxy resin with an epoxy equivalent of 270 and a total bromine content of 36wt%	Antimony trioxide	Carbon black	Carnauba wax	Type/Quantity
Example 39	0.7	2.3	1.0	0.3	0.3	V : 2.0
Example 40	0.7	2.3	1.0	0.3	0.3	II : 1.0
Example 41	0.7	2.3	1.0	0.3	0.3	IV : 2.0
Example 42	0.7	2.3	1.0	0.3	0.3	III: 2.0
Example 43	0.7	2.3	1.0	0.3	0.3	II : 3.0
Example 44	0.7	2.3	1.0	0.3	0.3	III: 2.0
Example 45	0.7	2.3	1.0	0.3	0.3	III: 1.0
Example 46	0.7	2.3	1.0	0.3	0.3	III: 1.0
Example 47	0.7	2.3	1.0	0.3	0.3	V : 2.0
Example 48	0.7	2.3	1.0	0.3	0.3	IV : 4.0
Example 49	0.7	2.3	1.0	0.3	0.3	II : 2.0
Example 50	0.7	2.3	1.0	0.3	0.3	I : 2.0
Example 51	0.7	2.3	1.0	0.3	0.3	V : 1.0
Comparative Example 17	0.7	2.3	1.0	0.3	0.3	IV : 4.0
Comparative Example 18	0.7	2.3	1.0	0.3	0.3	II : 3.0
Comparative Example 19	0.7	2.3	1.0	0.3	0.3	III: 2.0
Comparative Example 20	0.7	2.3	1.0	0.3	0.3	V : 2.0
Comparative Example 21	0.7	2.3	1.0	0.3	0.3	IV : 4.0
Comparative Example 22	0.7	2.3	1.0	0.3	0.3	I : 2.0
Example 52	0.7	2.3	1.0	0.3	0.3	0.0
Example 53	0.7	2.3	1.0	0.3	0.3	0.0
Example 54	0.7	2.3	1.0	0.3	0.3	0.0
Example 55	0.7	2.3	1.0	0.3	0.3	0.0

Table 15 Results of evaluation

	Heat resistance to solder	Reliability on moisture resistance
	Dipping in solder at 260 ℃ (Fraction defective)	PCT after dipping in solder at 260 ℃ (hr)
Example 39	0/16	310
Example 40	1/16	300
Example 41	0/16	320
Example 42	0/16	320
Example 43	0/16	300
Example 44	0/16	310
Example 45	2/16	320
Example 46	1/16	290
Example 47	0/16	340
Example 48	0/16	330
Example 49	0/16	350
Example 50	0/16	290
Example 51	2/16	300
Comparative Example 17	16/16	50
Comparative Example 18	14/16	90
Comparative Example 19	16/16	70
Comparative Example 20	16/16	60
Comparative Example 21	Melt-kneading was impossible ; evaluation impossible	
Comparative Example 22	10/16	110
Example 52	0/16	220
Example 53	4/16	190
Exampl 54	2/16	240
Example 55	2/16	230

Table 16 Results of evaluation

	Heat resistance to solder	Reliability on moisture resistance
	Dippingin solder at 260 ℃ (Fraction defective)	PCT after dipping in solder at 260 ℃ (hr)
Example 47	0/20	350
Example 51	0/20	350
Comparative Example 20	11/20	50

Claims

1. An epoxy resin composition for encapsulating a semiconductor device, which comprises
   (i) an poxy resin (A) comprising at l ast on f a bifunctional epoxy resin (a1) having a byphenyl skeleton and bifunctional eposy resin (a2) having a naphtal n skeleton,
   (ii) a curing agent (B), and
   (iii) a filler comprising a fused silica (C) consisting of 97 to 50 wt % of crushed fused silica (C1) of a mean particle diameter not more than 10 µm and 3 to 50 wt % of spherical fused silica (C2) of a mean particle diameter not more than 4 µm, wherein the mean particle diameter of the spherical fused silica is smaller than the mean particle diameter of the crushsed fused silica, and the amount of the filler is 75 to 90 wt% of the total weight of the composition.

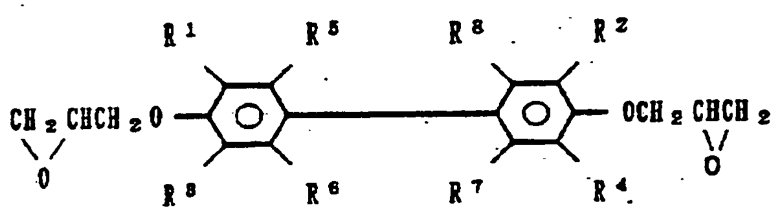
2. A composition according to claim 1, wherein the mean particle diameter of the crushed fused silica is less than 7 µm.

3. A composition according to claim 1 or 2, wherein the curing agent is a phenol type curing agent.

4. A composition according to claim 1,2 or 3, further containing a styrene type block copolymer (D).

5. A composition according to any preceding claim, further containing a copolymer (E) of (1) at least one compound selected from
   ethylene and α-olefin and (2) at least one compound selected from unsaturated carboxylic acid and derivate thereof.

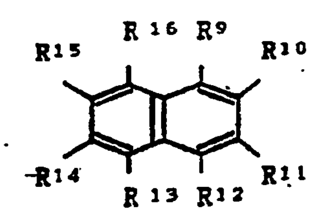
6. A compositon according to any preceding claim, wherein the bifunctional epoxy resin (a1) having a biphenyl skeleton is a compound represented by the following formula (I) :



......(I)

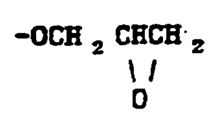
wherein R^1 to R^8 independently represent hydrogen atom, halogen atom or a lower alkyl froup having 1 to 4 carbon atoms.

7. A composition according to any preceding claim, wherein the epoxy resin bifunctional epoxy resin (a2) having a naphtalene skeleton is a compound represented by the following fomula (II):



.......(II)

wherein two of R^9 to R^{16} independently represent a group represented by



and those remaining independently represent hydrogen atom, halogen atom or a lower alkyl group having 1 to 4 carbon atoms.

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(54) **An epoxy resin composition for encapsulating a semiconductor device.**

(57) An epoxy resin composition for encapsulating a semiconductor device comprises (i) an epoxy resin (A) comprising at least one of a bifunctional epoxy resin (a1) having a biphenol skeleton and a bifunctional epoxy resin (a2) having a naphthalene skeleton, (ii) a curing agent (B), and (iii) a filler comprising fused silica (C) consisting of 97 to 50 wt% of crushed fused silica (C1) of a mean particle diameter not more than 10 μm and 3 to 50 wt% of spherical fused silica (C2) of a mean particle diameter not more than 4 μm, wherein the mean particle diameter of the spherical fused silica is smaller than the mean particle diameter of the crushed fused silica, and the amount of the filler is 75 to 90 wt% of the total weight of the composition.

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DOCUMENTS CONSIDERED TO BE RELEVANT

Category	Citation of document with indication, where appropriate, of relevant passages	Relevant to claim	CLASSIFICATION OF THE APPLICATION (Int. Cl.5)
Y	CHEMICAL PATENTS INDEX, BASIC ABSTRACTS JOURNAL Section Ch, Week 8950, 14 February 1990 Derwent Publications Ltd., London, GB; Class A, AN 89-367658 & JP-A-01 275 620 (SUMITOMO BAKELITE KK) 6 November 1989 * abstract *	1-4,6	C08G59/24 C08K3/36 C08L63/00 C08L31/00 C08L53/00 H01L23/29
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A	EP-A-0 142 825 (PHILLIPS PETROLEUM) 29 May 1985 * page 3, line 11 - line 14; claim 1 *	5	C08G C08K H01L

The present search report has been drawn up for all claims

Place of search	Date of completion of the search	Examiner
THE HAGUE	15 NOVEMBER 1991	O'SULLIVAN T.P.

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